Solid-state anaerobic digestion of lignocellulosic biomass: Recent progress and perspectives

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Abstract

Solid-state anaerobic digestion (SS-AD) has gained popularity in the past decade as an environmentally friendly and cost-effective technology for extracting energy from various types of lignocellulosic biomass. According to data of biomass and methane yields of lignocellulosic feedstocks, crop residues have the highest methane production potential in the U.S., followed by the organic fraction of municipal solid waste (OFMSW), forestry waste, and energy crops. Methane yield and process stability of SS-AD can be improved by different strategies, such as co-digestion with other organic wastes, pretreatment of lignocellulosic biomass, and optimization of operating parameters. Different models for SS-AD have been developed, and insights into SS-AD processes have been obtained via microbial community analysis, microscope imaging, and tracer techniques. Future research and development in SS-AD, including feedstock identification and co-digestion, feedstock storage and pretreatment, SS-AD reactor development, digestate treatment, and value-added production, were recommended.

Keywords: Solid-state anaerobic digestion; Lignocellulosic biomass; Waste management; Methane; Biogas production

1. Introduction

More than 340 million dry tonnes of lignocellulosic biomass, such as crop residues and forest residues, were generated in the U.S. in 2012 (Balan, 2014). Utilization of this waste biomass as a renewable feedstock to produce energy and valuable products will not only improve the sustainability of agricultural and forestry systems, but also alleviate our dependence on fossil fuels and reducing greenhouse gas emissions.

Anaerobic digestion (AD) is a plausible dual-purpose technology for treating complex biomass wastes and converting organic matter into biogas, which mainly consists of methane (CH_4) and carbon dioxide (CO_2) with traces of other impurities, such as hydrogen sulfide (H_2S), ammonia (NH_3), and water vapor. Biogas has been used to produce electricity using combined heat and power (CHP) systems. Purified methane from biogas has also been upgraded to compressed 'natural' gas (Bio-CNG) or liquefied 'natural' gas (Bio-LNG) for transportation fuels (Yang et al., 2015). Raw or purified biogas can also be upgraded into liquid fuels via biological or thermochemical conversion methods (Sheets et al., 2015; Yang et al., 2015). The digestate contains large amounts of nutrients (e.g. nitrogen and phosphate) and can be used as a fertilizer (Li et al., 2011a; Yang et al., 2015).

AD can be operated at different total solids (TS) contents. Most large-scale AD processes are operated at TS contents of less than 15% and are classified as liquid-AD (L-AD), which is suitable for treating high moisture wastes, such as sewage sludge, food wastes, and animal manures (Li et al., 2011a). However, L-AD requires a large amount of water for treating low moisture feedstocks, such as lignocellulosic biomass, leading to decreased volumetric methane productivity and a huge volume of wastewater to be treated. In contrast, solid-state AD (SS-AD) generally operates at TS contents of more than 15% and is more suitable to digest lignocellulosic

biomass (Li et al., 2011a; Yang et al., 2015). Compared to L-AD, SS-AD has several advantages, such as higher volumetric methane productivity, reduced energy requirements for heating, less wastewater generation, and a low-moisture digestate that is easier to handle (Li et al., 2011a; Xu et al., 2014b). However, SS-AD usually has an inadequate mass transfer and high organic loading, which result in reduced methane yields and a higher tendency to accumulate inhibitors, such as ammonia and volatile fatty acids (VFAs), than L-AD (Motte et al., 2013b; Xu et al., 2015). To date, L-AD has been well-established in many regions of the world, while SS-AD systems are more commonly found in European countries but not in others such as the U.S. (Yang et al., 2015).

The number of publications regarding SS-AD rose sharply between 2010 and 2014 according to a Google Scholar search result (Figure 1). Increasing efforts have been made to optimize the SS-AD process, and achievements have been made in improving methane yield, stability, and benefit-to-cost ratio. This review focuses on the research progress in SS-AD of lignocellulosic biomass that has been reported during the last decade, including aspects of feedstocks, operational strategies, and in-depth insights. Several perspectives for further research in SS-AD of lignocellulosic biomass are also discussed.

2. Feedstocks for solid-state anaerobic digestion

2.1. Recently evaluated feedstocks

Corn stover, including all the above-ground parts of the corn plant (stalks, leaves, tassels, cobs and husks, except for the grain) comprises more than three-fourths of the total crop residues that are produced in the U.S. For every 1 dry tonne of corn grain produced, about 0.94 dry tonnes of corn stover is produced. Although some of the corn stover needs to remain in the field to

maintain soil quality, about 75 million dry tonnes are available for harvesting in the U.S. each year (Ertl, 2013). Methane yields of 130-239 L/kg volatile solids (VS) were obtained by 30-day SS-AD of corn stover at TS contents of 18-22% and temperature of 37°C, depending on different inocula and feedstock-to-inoculum (F/I) ratios (Brown et al., 2012; Xu and Li, 2012; Xu et al., 2013).

Wheat straw, the above-ground biomass remaining after wheat harvest, is another abundant crop residue. It was estimated that around 33 million dry tonnes of wheat straw are available in the U.S. each year (Cardona et al., 2009). SS-AD of wheat straw and spent wheat straw that has been used for livestock bedding has been investigated at a TS content of 22% and F/I ratios of 2, 4, and 6 (based on VS) (Cui et al., 2011). The highest methane yield of wheat straw and spent wheat was about 96 and 150 L/kg VS, respectively, during 30 days of SS-AD. For both feedstocks, similar methane yields were obtained at F/I ratios of 2 and 4, while all digesters failed at the F/I ratio of 6 (Cui et al., 2011). Higher methane yields up to 200 L/kg VS were obtained by SS-AD of wheat straw at a TS content of 20% and F/I ratio of 2 (Y. Lin et al., 2014). Since the same particle size (10 mm screen) of wheat straw was used in the two studies, the difference in methane yields may be due to differences in the inocula and sources of the feedstock.

Yard trimmings are a major component of the organic fraction of municipal solid waste (OFMSW), and contain mainly wood chips with small amounts of grass and leaves. Each year, about 31 million dry tonnes of yard trimmings are generated in the U.S. (USEPA, 2015). In 2013, about 19 million tonnes of yard trimmings were recycled for composting, during which the inherent energy was wasted as respiration heat (USEPA, 2015). Alternatively, these yard

92 trimmings can be used for methane production via SS-AD, while the digestate can still be used
93 as a fertilizer or soil amendment.

94 *Forestry waste* includes materials from operations within forest and timberland areas and
95 residues generated during processing for wood products, such as lumber, paper, and furniture.
96 The waste biomass could be converted to bioenergy and bioproducts (Walsh, 2008). The total
97 logging residues generated in 2002 in the U.S. was 46.3 million tonnes, including 30.7 million
98 tonnes of hardwoods and 15.6 tonnes of softwoods (Walsh, 2008). However, methane yields
99 from untreated yard trimmings were relatively low (20-74 L/kg VS during 30 days of SS-AD),
100 due to the recalcitrant woody structure (Ge et al., 2015, 2014; Y. Lin et al., 2014; Zhao et al.,
101 2014a, 2014b). A higher methane yield (97 L/kg VS) was obtained by increasing the period of
102 SS-AD to 60 days (Y. Lin et al., 2014). Forestry woody biomass also showed low methane yields
103 during SS-AD, especially for softwoods which have high lignin contents (Brown et al., 2012).

104 *Energy crops*, such as switchgrass, miscanthus, and giant reed, have been increasingly
105 studied in recent years as reliable and sustainable biomass feedstocks with high biomass yields
106 and low production costs (Corno et al., 2014). These energy crops can adapt to different climate
107 and soil conditions, and require low fertilizer inputs. Their high water and nitrogen use efficiency
108 enables them to grow on marginal land; thus, they do not compete with food and feed production.
109 About 222,000 ha of marginal land in ten Midwestern U.S. states was identified as suitable for
110 energy crop production (Gelfand et al., 2013; Izaurralde and Zhang, 2013). According to Corno
111 et al. (2014), the biomass yields for switchgrass, miscanthus, and giant reed were about 15, 22,
112 and 45 tonnes per ha per year, respectively. If 20% of the marginal land was used for growing
113 each of the energy crops, about 3.3 tonnes of switchgrass, 4.9 tonnes of miscanthus, and 10.0
114 tonnes of giant reed could be produced each year. Methane yields of these energy crops were

comparable to those of crop residues, and higher than those of woody biomass (Table 1). The methane yield of switchgrass was 113-127 L/kg VS at 35°C-37°C, and 145-167 L/kg VS at 55°C (Brown et al., 2012; El-Mashad, 2013; Sheets et al., 2015). *Miscanthus sinensis* harvested in fall and spring showed methane yields of about 130 and 170 L/kg VS for 30-day and 60-day SS-AD, respectively (Vasco-Correa and Li, 2015). In addition, reported methane yields by 30-day SS-AD of giant reed were about 100-147 L/kg VS (Liu et al., 2015a; Yang and Li, 2014).

Although higher methane yields of lignocellulosic biomass could be achieved using L-AD, the improvement varied dramatically depending on feedstocks. For example, methane yield from pine during L-AD was more than 3 times as high as that in SS-AD, while methane yields by L-AD of wheat straw, leaves, yard waste, maple or giant reed were only 8-22% higher than those by SS-AD (Brown et al., 2012; Yang and Li, 2014). SS-AD of corn stover and switchgrass even obtained slightly higher (5-6%) methane yields than L-AD (Brown et al., 2012). Comparisons of the methane production potential by SS-AD of different feedstocks in the U.S. were conducted based on methane yields and biomass production and are shown in Table 1. Generally, crop residues showed higher methane production potential than OFMSW, forestry waste, and energy crops. Corn stover had the highest methane production potential of 17,010 million m³/year followed by wheat straw (6,121 million m³/year) and yard trimmings (2,383 million m³/year). Forestry hardwood residues and giant reed showed comparable methane production potentials of about 1,400 million m³/year. It should be noted that the methane production potential for different regions may vary with climate and available plant species.

2.2. Solid-state co-digestion of lignocellulosic biomass with other feedstocks

Carbon to nitrogen (C/N) ratio is a crucial factor that affects the AD performance.

Lignocellulosic biomass may obtain low methane yields during mono-digestion due to their high C/N ratios (usually higher than 50). Solid-state co-digestion of lignocellulosic biomass with feedstocks that have lower C/N ratios has been used to obtain a proper C/N ratio (generally lower than 20), and proved reliable for improving the methane yield (80%-691%) (Brown and Li, 2013; L. Lin et al., 2015; Y. Lin et al., 2014; Mussoline et al., 2013; Xu and Li, 2012; Zhu et al., 2014) (Table 2).

Besides, the performance of co-digestion also varied with other operation parameters, such as feedstock-to-inoculum (F/I) ratio. Methane yield of solid-state co-digestion of hay and soybean processing waste decreased when the F/I ratio increased from 1 to 5 (Zhu et al., 2014). Similarly, solid-state co-digestion of spent mushroom substrate and yard trimmings (1:1) failed when the F/I ratio was higher than 2 (Y. Lin et al., 2014). The main reason was believed to be VFA accumulation that caused pH drop and consequently inhibited the methanogenesis process. During SS-AD of lignocellulosic biomass, buffer capacity is generally provided by the inoculum. As a result, SS-AD processes at higher F/I ratios were less stable than those at lower F/I ratios when VFA accumulation occurred, thus showed decreased methane yields or even failed.

2.3. Pretreatment of lignocellulosic biomass

Various pretreatment technologies, such as dilute acid, steam explosion, alkaline, hot water, wet oxidation, and biological methods, have been used to improve methane yield during AD of different types of lignocellulosic biomass, including food processing wastes, agricultural residues, municipal solid waste, and energy crops (Figure 2a and 2b) (Yang et al., 2015; Zhang et al., 2011;

Zhao et al., 2014b; Zhu et al., 2010). In general, improved methane production was obtained, although the increase of methane yields varied dramatically with feedstock and pretreatment methods. Alkaline pretreatment was particularly effective for softwood, such as spruce which showed a low methane yield (30 L/kg VS) due to its high lignin content (29%) (Mohsenzadeh et al., 2012). In a study of alkaline pretreatment with different reagents (NaOH/thiourea, NaOH/urea, NaOH/urea/thiourea, and NaOH/PEG) at different temperatures (-15°C, 0°C, 22°C, and 80°C), about 11%-23% of lignin in spruce was removed in 16 h of pretreatments. The pretreatments improved glucose yield of spruce from 11% to 27%-57% (Mohsenzadeh et al., 2012). The highest glucose yield (57%) was obtained by pretreatment with NaOH/thiourea at -15°C which increased the methane yield to 210 L/kg VS during the subsequent AD process (Mohsenzadeh et al., 2012). White rot fungus, *Ceriporiopsis subvermispota*, has been commonly used for biological pretreatment of lignocellulosic biomass. A 400% increase in methane yield from Japanese cedar wood chips was achieved using *C. subvermispota* for fungal pretreatment, although the improved final methane yield was still low (43 L/kg VS). Fungal pretreatment of albizia chips with *C. subvermispota* achieved a 370% increase in methane yield, which was more effective than pretreatment of yard trimmings with the same strain (Ge et al., 2015). After pretreatment, most lignocellulosic biomass has been shown to obtain methane yields higher than 200 L/kg VS during SS-AD (Figure 2a).

For several types of lignocellulosic biomass, more than one type of pretreatment method has been studied, allowing for comparison of methods. For example, steam explosion and alkaline pretreatment have each been tested for improving methane production by SS-AD of hardwood, and achieved comparable methane yields (Figure 2a) (Yang et al., 2015). Hot water pretreatment obtained a 222% increase in methane yield from rice straw, which was higher than

that obtained using alkaline pretreatment (Figure 2b) (Yang et al., 2015). Alkaline pretreatment seems more effective than steam explosion and hot water in improving methane yield from wheat straw (Figure 2b) (Yang et al., 2015). However, it should be noted that these data were obtained from different studies. Further studies with side-by-side comparisons between different pretreatment methods using the same feedstock are needed in order to determine which method is more effective for a specific lignocellulosic biomass. Besides, most of the above-mentioned pretreatment methods were evaluated for L-AD, and their performance needs to be further examined for SS-AD.

3. Operational strategies for solid-state anaerobic digestion

3.1. Inoculation

3.1.1. Inoculum source and composition

For traditional commercial SS-AD digesters, the finished solid material, or “digestate”, is commonly recycled as an inoculum. Recent research have shown that the finished material (effluent) from L-AD can be a good inoculum for SS-AD (Cui et al., 2011; Xu et al., 2013). Compared to other inocula, such as rumen fluid, manure, lake sediments, sewage sludge, and SS-AD digestate, L-AD effluent has been found to more quickly initiate a SS-AD processes (Xu et al., 2016; Yang et al., 2015). Based on this concept, a patented technology (Li et al., 2014) has recently been applied in a pilot-scale digester in Zanesville, OH, USA, where the solid and liquid digesters are built side-by-side, and the effluent from liquid digesters is used as inoculum for the solid-state digester.

The source and composition of the inoculum have been found to significantly influence the methane yield, startup time, and stability of SS-AD. Traditional studies based on L-AD, in which

methanogenesis is usually the rate limiting step, have mainly focused on the population of methanogens in the inoculum. However, with the increasing popularity of SS-AD of lignocellulosic biomass, more researches have started to emphasize the importance of hydrolytic microbes in the inoculum for methane production. Xu et al. (2013) reported that using dairy manure digester effluent as inoculum could obtain about 30% and 100% higher methane yields from corn stover compared to using effluent from food waste and sewage sludge digesters, respectively. This result could be attributed to the higher populations of hydrolytic microbes in dairy manure effluent (Xu et al., 2013). Similarly, Gu et al. (2014) found that the highest methane yield of rice straw was obtained by using digested dairy manure as inoculum, which had higher cellulase and xylanase activities and more balanced nutrients than other inocula studied. Ma et al. (2013) quantitatively determined that the ratio of hydrolytic microbes to methanogens should be below 24, at which hydrolysis became the rate limiting step and VFA accumulation would be avoided. However, most of these studies did not collect microbial community and degradation data during the SS-AD process. Further experiments should be conducted to collect direct evidence for the effect of initial microbial compositions in inoculum on the subsequent SS-AD.

3.2.2. Inoculation size

Inoculation size has been proposed to be one of the most important factors in batch SS-AD to achieve high methane yield and digester stability. Larger inoculation size is usually preferred to provide more methanogens for the fast and successful startup of SS-AD (Yang et al., 2015). A wide range of F/I ratios has been studied in SS-AD of lignocellulosic biomass. F/I ratios ranging from 2 to 3 (VS basis) have been shown to be relatively robust for SS-AD under mesophilic

temperatures (Xu and Li, 2012; Zhu et al., 2014). At thermophilic temperatures, a relatively high F/I ratio was preferred. For instance, the highest biogas yield of corn stover in mesophilic SS-AD was obtained with an F/I ratio of 2.43 (VS basis), while the highest yield under thermophilic conditions was achieved at an F/I ratio of 4.58 (Li et al., 2011b). The optimal F/I ratio for SS-AD of yard trimmings was reported to be 1 at mesophilic temperatures (Brown and Li, 2013), but 4–6 at thermophilic conditions (L. Lin et al., 2014). With the addition of buffers, higher F/I ratios of 30-50 were shown to be effective for SS-AD, but the process required 300 days which were about 5-10 times longer than SS-AD with low F/I ratios (Motte et al., 2013a).

More recent studies show that inoculation size not only affects microbial population, but also physical and chemical properties of SS-AD. In some research, SS-AD with low F/I ratios obtained higher methane yield partially because of the additional moisture content provided by the inoculum, which is beneficial to the mass transfer and microbial growth (L. Lin et al., 2014)(Xu et al., 2016; Yang et al., 2015). Using finished AD materials as inoculum usually provides additional buffering capacity to the subsequent SS-AD process, as well as nitrogen and micro-nutrients (Gu et al., 2014; Shi et al., 2013). These influences should be considered in SS-AD research and system design. Statistical models indicated that inoculation size also had interactions with other SS-AD operating parameters, such as lignin content, TS content, and particle size of the feedstock (Motte et al., 2013a; Xu et al., 2014a). Motte et al., (2013a) also found the influence of inoculation size to be more dominant during the initial stage of AD (e.g., the first 10-30 days within the 300-day SS-AD), but less significant at the later stages.

3.2. *Partial premixing/ leachate recirculation*

Due to the high TS and viscosity in SS-AD, continuous and intensive mixing is usually difficult during the digestion process. Therefore, an initial premixing of feedstock with inoculum prior to loading into the digester is required. In large-scale digesters, an uneven distribution of inoculum is expected to occur due to the incomplete premixing. Zhu et al. (2014) simulated this situation at lab-scale, and found that at an F/I ratio of 3, partially premixing 50% of the inoculum with the feedstock and adding another 50% to the top of the digester achieved almost the same methane yield as the reactors with complete premixing of feedstocks and inoculum. The inoculum added from the top of the digester gradually permeated through the feedstock and successfully initiated the SS-AD process. However, the startup time was much longer than the completely premixed reactors (Zhu et al., 2014).

Incomplete premixing has also been used to reduce the inoculation size and recover acidified SS-AD digesters. SS-AD of corn stover usually requires an F/I ratio of less than 4 to prevent acidification (Shi et al., 2014; Xu et al., 2014a). However, by mixing a portion of the inoculum with the feedstock, and then adding the remaining inoculum as two layers (Figure 3), SS-AD of corn stover can be operated at a high F/I ratio of 6 (Zhu et al., 2015). This is because partial mixing limited the fast mass transfer of the excessive VFAs to the methanogens, thus prevents the acidification of the inoculum. However, compared with complete premixing of feedstock and inoculum with low F/I ratios, the methane yields and methane production rate obtained from partial premixing and high F/I ratio were significantly lower, also due to the slow mass transfer of VFAs to methanogens (Zhu et al., 2015). The researchers also found that for acidified digesters with an F/I ratio of 10, reducing the overall F/I ratio to 4 by adding extra inoculum on the top successfully recovered the failed digesters. More improvements on reactor

designs are still required to overcome the mixing problem in SS-AD and improve the efficiency of the feedstock utilization and methane production rate.

Leachate recirculation is a commonly used method to enhance mass transfer in SS-AD by redistributing substrates, nutrients, and microorganisms (André et al., 2015). However, leachate recirculation did not obtain significant increases of biogas production in one SS-AD study (Zhu et al., 2014). Another study found that leachate recirculation only improved methane yield at the early stage of AD, but no effect on methane production rate after 19 days. The authors attributed this result to the physical structure of the manure during the AD process, which reduced the liquid percolation through the reactor (André et al., 2015). Currently, experimental study and optimization of leachate recirculation in SS-AD are relatively limited. However, it appears that leachate recirculation, although theoretically beneficial for SS-AD, needs precise control to achieve positive effects.

3.3. Total solids content

Increasing the TS content in SS-AD can reduce the digester volume and capital cost. However, the methane yield (volume of methane produced per unit VS of substrate, $\text{L/kgVS}_{\text{feed}}$) and methane production rate ($\text{L/kgVS}_{\text{feed}}/\text{day}$) tend to decrease as TS increases above a threshold of about 15%-20% (Xu et al., 2014b). Fernández et al. (2010) reported a 17% decrease in organic waste degradation when the TS content increased from 20% to 30%. Brown et al. (2012) reported reduced methane yields and cellulose hydrolysis rates of lignocellulosic biomass in SS-AD compared to those in L-AD.

Although the increase of TS content has been commonly observed to reduce substrate degradation and biogas yield, not much explanation or evidence has been provided for the

mechanisms behind it. Recently, more studies have been conducted to explain the effect of TS content on methane yield, and most of them attribute the reason to mass transfer (Table 3). Generally, three types of explanations are given: 1) diffusion limitation caused the accumulation of different inhibitors; 2) diffusion limitation reduced microbial access to the substrate; and 3) reduced water content directly affected microbial metabolism. Some of these hypotheses are supported by direct experimental data. For example, Abbassi-Guendouz et al. (2012) hypothesized that limited liquid-gas mass transfer caused H₂ and CO₂ accumulation at high TS contents and inhibited methanogens, and an experimental measurement of H₂ and CO₂ partial pressures in AD have been used to verify this hypothesis (Cazier et al., 2015). However, most of the hypotheses listed in Table 3 have only been verified by mathematical models using indirect data such as pH values and methane production, but have not been supported by direct experimental data (Xu et al., 2015). More experimental evidence is needed to verify these hypotheses, and deepen our understanding of SS-AD processes.

3.4. Temperature

Both mesophilic (around 37°C) and thermophilic (around 55°C) conditions have been used for SS-AD. Mesophilic AD is considered more stable and better established than thermophilic AD (Yang et al., 2015). However, thermophilic AD usually shows a better startup performance than mesophilic AD due to an accelerated hydrolysis process, the major rate-limiting step during AD (Yang et al., 2015).

It was found that VFA generation and feedstock degradation were faster in the thermophilic digester than in the mesophilic digester, and that 95% of the methane yield potential was achieved in 11 and 27 days under thermophilic and mesophilic conditions, respectively (Hegde

and Pullammanappallil, 2007). One concern about thermophilic SS-AD is that enhanced activity of fermentative microbes in thermophilic conditions may cause accumulation of VFAs and result in inhibitory effects on the AD process, which may vary depending on several factors such as TS content, F/I ratio, and C/N ratio (L. Lin et al., 2014; Shi et al., 2014, 2013; Yan et al., 2015). The inhibitory effect could be mitigated by increasing the buffer capacity of SS-AD systems. For example, Hegde and Pullammanappallil (2007) flooded a bed of vegetable waste and wood chips in mesophilic (38°C) and thermophilic (55°C) SS-AD systems with leachate from previous digestion of these feedstocks to provide adequate alkalinity.

Another concern about thermophilic SS-AD is the high energy input for heating, which may offset its higher methane production yields and rates (Li et al., 2011a; Sheets et al., 2015). Moreover, the energy input required to maintain thermophilic temperatures may vary significantly for different seasons. Therefore, it is important to evaluate the performance of SS-AD at different temperatures based on net energy production. For example, Sheets et al. (2015) compared the performance between thermophilic and mesophilic SS-AD of switchgrass. Although thermophilic SS-AD showed greater methane yield than mesophilic SS-AD, mesophilic conditions had higher net energy production than thermophilic conditions due to reduced initial heating requirements after inoculation (Sheets et al., 2015). Currently, few studies have been reported on net energy production by mesophilic and thermophilic SS-AD.

4. In-depth insights into solid-state anaerobic digestion processes

4.1. Mathematical modeling

Modeling is believed to be an effective approach of understanding reaction mechanisms, predicting system performance, and facilitating process design (Fdez-Güelfo et al., 2012).

Different SS-AD models developed in the past decades have been be classified into theoretical, empirical, and statistical models (Xu et al., 2015). Theoretical models can provide more insight into the complex system mechanisms, but need a series of inputs and thus are usually too complicated for general applications (Weinrich and Nelles, 2015). In comparison, empirical models can be less complicated, while some of their assumptions and equations might be developed empirically without theoretical foundations. Statistical models can be easily derived from the correlations between data, but are sometimes “black or grey boxes” that are hard to interpret.

Most of the SS-AD theoretical models published before 2005 mainly focused on the effect of the heterogeneous initial distribution of inoculum in the substrate, which caused VFA accumulation in some regions of the reactor (Table 4). Based on model simulation results in these studies, highly dispersed inoculum, vigorous mixing, and leachate recirculation were unfavorable in SS-AD, because under these conditions, the inoculum particles were more likely to be acidified by the VFAs (Vavilin, 2004).

Since 2011, several new SS-AD theoretical models have been developed that build on previous knowledge. Three recent studies derived their SS-AD models based on the Anaerobic Digestion Model No.1 (ADM1), the most comprehensive model for L-AD, and assumed a homogenous system with complete mixing inside (Abbassi-Guendouz et al., 2012; Bollon et al., 2011; Liotta et al., 2015). In contrast, another research is more similar to those previous SS-AD models published before 2005, which assumed a heterogeneous distribution of inoculum within the SS-AD digesters (Xu et al., 2014b).

These recent SS-AD models have focused on the interpretation of the effect of TS content on methane yield and production rate (Table 4). Most of these models assumed that TS content is

a key parameter that affects the mass transfer between the gas-liquid-solid phases in SS-AD, and in turn affects the rate of substrate degradation and accumulations of inhibitors. For example, TS content has been assumed to affect the hydrolysis rate constant (Abbassi-Guendouz et al., 2012; Liotta et al., 2015), maximum microbial growth rate or half saturation coefficient (Fernández et al., 2010; Liotta et al., 2015; Pommier et al., 2007), and diffusion coefficient (Abbassi-Guendouz et al., 2012; Xu et al., 2014b) in SS-AD. Two recently published statistical models also statistically calculated how TS content affect methane production in SS-AD (Motte et al., 2013a; Xu et al., 2014a).

To date, there is no universal consensus of the best modeling approach for SS-AD (Xu et al., 2015). Although significant progress has been made in understanding SS-AD processes, reaction mechanisms in SS-AD, such as the influences of TS content, mass transfer, substrate composition, temperature, and heat transfer, are still not clear (Li et al., 2011a; Xu et al., 2015). Further investigation of SS-AD mechanisms is essential for the development of SS-AD mathematical models.

4.2. Microbial community dynamics

The first published study of microbial dynamics during SS-AD compared the microbial communities in SS-AD of corn stover at mesophilic and thermophilic conditions using most probable number (MPN) and denaturing gradient gel electrophoresis (DGGE) analyses (Shi et al., 2013). The populations of cellulolytic and xylanolytic microbes detected in thermophilic systems were about 10–50 times greater than those in mesophilic ones, corresponding to the faster and greater reductions of cellulose and hemicellulose in thermophilic digesters during the first 12 days compared to those in mesophilic SS-AD. Using similar methods, the effect of inoculation

ratio on microbial community dynamics in SS-AD were also investigated (Shi et al., 2014). Non-microbial factors of the inoculum, such as alkalinity, were found to be more decisive on the final methane yield of corn stover, while the population of methanogens affected the kinetics of VFA consumptions and methane production. In a follow up researches, the microbiome composition and their temporal succession were determined in thermophilic and mesophilic SS-AD, as well as acidified SS-AD reactors using Illumina sequencing of 16S rRNA gene amplicons (Li et al., 2016, 2015). The genus *Methanothermobacter* dominated the archaea in the thermophilic SS-AD reactors, while *Methanoculleus* dominated in the mesophilic reactors (Li et al., 2015). Syntrophic acetate oxidation coupled with hydrogenotrophic methanogenesis was found an important pathway for biogas production during thermophilic SS-AD (Li et al., 2015), and *Methanomassiliicoccus* abundance was positively correlated to daily biogas yield in the mesophilic SS-AD process (Li et al., 2016).

Microbial community analysis has also been employed to study the effect of inhibitors in SS-AD. The inhibitive effect of total ammonia nitrogen (TAN) (including NH_3 and NH_4^+) on mesophilic SS-AD of corn stover was investigated in batch reactors using a culture-based method, in which cellulolytic microbes and acetotrophic methanogens were observed to be more sensitive to TAN than xylanolytic microbes (Wang et al., 2013). However, culture-based enumeration is generally believed to be more time and labor consuming, less informative and accurate than those PCR-based enumerations (Yue et al., 2013). The inhibition of acetate in SS-AD of food waste was evaluated by DGGE, and the acetyl-CoA enzyme was measured to reflect the activity of acetotrophic methanogens (Xu et al., 2014a). Significant changes of the dominant species, including *Methanosarcina mazei*, *Methanobacterium* sp., and uncultured *Methanocorpusculum* sp., were observed with increased acetate concentration. Acetotrophic

methanogenesis was found to be more sensitive to the increased organic loading than hydrogenotrophic methanogenesis; the imbalance between these two metabolic pathways would lead to low methane production.

Generally speaking, studies about microbial community dynamics in SS-AD are relatively rare compared to the numerous ones in L-AD. One reason is that SS-AD is still an emerging research area compared with L-AD, and another might be the difficulty of taking samples during the SS-AD process due to labor-consuming, destructive sampling methods. In the existing studies, discrepancies in SS-AD microbial communities were found. The dynamics of microbial community changes during SS-AD are complex, and many factors, such as substrate, inoculum source, operating conditions, and sampling procedures, may affect the results.

4.3. Ultrastructural observation of process dynamics

New experimental instrumentation has been used to illustrate the physicochemical and biological processes in SS-AD of lignocellulosic biomass at microscale. For example, currently research about structural changes of lignocellulosic biomass during SS-AD is still relatively simple and usually only the losses of cellulose and hemicellulose are reported. One research utilized transmission electronic microscopy to observe the ultrastructural changes during wheat straw degradation in SS-AD (Motte et al., 2015). The images of the plant cell wall structure and the distribution of microbes showed that the microorganisms degraded wheat straw from the central to the outer tissue (i.e. parenchyma to epidermis). Mass transfer is another aspect in SS-AD which requires more advanced detection methods for in-depth study. In one research, potassium iodide trace was used to observed the solute diffusion in the solid medium of SS-AD (Bollon et al., 2013). The value of diffusion coefficient in SS-AD was measured to be 200 times

smaller than that in pure water ($1.98 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$). However, the diffusion of inorganic salts can be very different from the actual organic substances, such as sugars, VFAs, enzymes, and microbes. The direct measurement of mass transfer coefficients of those intermediate products in SS-AD can be challenging as they are bioconvertible. In future research, organic and non-biodegradable tracers, or isotope-labeled organic compounds, can be used to determine the degradation and mass transfer processes during SS-AD (Xu et al., 2015).

The diverse experimental methods used in L-AD systems can be applied to SS-AD to gain further insight on the process and provide direct evidence to support SS-AD hypotheses, such as the rate limiting step, heterogeneous distribution of microbial consortia, and changes of substrate metabolic rate constants at different operating conditions. It should be noted that due to the solid nature of the SS-AD medium, established analytical techniques used for solid systems, such as soil, rumen, compost, and solid-state fungi pretreatment and fermentation, can also be adopted as useful experimental tools for the verification of SS-AD model assumptions.

5. Perspectives on solid-state anaerobic digestion

Despite increasing numbers of publications in recent years, SS-AD of lignocellulosic biomass is still not as well understood and widely applied as L-AD. Future research should focus on the fundamental study of SS-AD mechanisms, such as mass transfer and microbial community dynamics, in order to better control the process and enhance biogas yield (Sawatdeenarunat et al., 2015; Yang et al., 2015). The next steps in SS-AD research should also include the transition of current lab-scale SS-AD research towards commercialization. Further research and development could be devoted to the entire process chain, including new feedstock

identification, storage, pretreatment, AD process design, post-treatment of digestate, and the evaluation of benefit-to-cost ratios and environmental impacts (Figure 4).

5.1. Feedstock identification and co-digestion

Co-digestion is commonly needed in commercial scale digesters to increase the methane yield and to meet the feedstock handling capacity of more than 20,000 tonnes per year (Pöschl et al., 2010). In Europe, crop silages are the most widely used substrates for SS-AD (Kalamaras and Kotsopoulos, 2014), and crop residues have been identified as having the highest potential for biogas production in the U.S. (Table 1). For scale-up and wide application of SS-AD of lignocellulosic biomass, identification and conversion of a variety of untraditional and locally abundant biomass wastes may be required in order to reduce transportation costs and the dependence on a single feedstock, which may be affected by factors such as geographical location and season. For example, papaya and sweet potato production waste, and albizia leaves and chips obtained from invasive species removal, have been identified as potential feedstocks in Hawaii (Ge et al., 2014). In Ohio, expired pet food, soybean processing waste, and spent mushroom culture media have been investigated as alternative feedstock sources to co-digest with crop residues in SS-AD (Y. Lin et al., 2015, 2014; Xu and Li, 2012; Zhu et al., 2014). In southern Europe climates, where water is relatively scarce, sorghum, cardoon, and milk thistle crops are considered highly promising for biogas production (Kalamaras and Kotsopoulos, 2014). More research is needed to identify new locally available and low-cost feedstocks for SS-AD, and to develop co-digestion strategies to utilize these resources.

5.2. Evaluation of feedstock storage and pretreatment methods for SS-AD

Storage of biomass feedstocks is another crucial aspect for maintaining a viable biofuel supply chain. Further improvements of storage methods are necessary to reduce dry matter loss and storage costs, and to increase the biomass digestibility. Ensilage has been recently evaluated as a promising method for wet storage and simultaneous pretreatment for SS-AD of energy crops. Ensilage is commonly used for preserving wet feedstocks in the livestock industry. It relies on lactic acid bacteria in the biomass to ferment free sugars to organic acids and ethanol. The organic acids, especially lactic acid, reduce pH to about 4, thus preventing degradation of biomass by inhibiting the activity of microorganisms. Ensiled biomass is particularly suitable for AD since these organic acids can be readily utilized for biogas production. Urea and food waste have been investigated as additives during ensilage, and were found to increase the lignin degradation and enhance methane yield of SS-AD (Liu et al., 2015a, 2015b). Ensilage of various feedstocks, such as corn stover and energy crops, has been studied for methane production via L-AD (Liu et al., 2015a). Future studies on the ensilage of SS-AD feedstocks, including the process conditions, influence of harvest method, harvest time, and different additives, are needed.

Many physical, chemical and biological pretreatment methods have been investigated for improving the methane yield from lignocellulosic biomass, but only a few of them have been tested for SS-AD (Zheng et al., 2014). Different from L-AD, SS-AD requires high-TS feedstocks, thus pretreatment conditions need to be adjusted to avoid the generation of large amounts of leachate and wastewater. In addition, the high organic loading of SS-AD limits the input of large amounts of easily digestible materials, and thus the extent of pretreatment required for SS-AD needs to be experimentally optimized (Zhu et al., 2010), or co-substrates that can dilute the produced VFAs or increase alkalinity should be evaluated. As mentioned in section 2.3, side-by-

side comparison between pretreatment methods using the same feedstock as well as an overall evaluation of the process economics and environmental impacts are needed.

5.3. Development of reactor systems for commercial-scale SS-AD

Batch reactors with leachate recirculation, such as Bekon, and continuous reactors with different agitations, such as Dranco, Kompogas, and Valorga, are two main types of SS-AD systems (Li et al., 2011a). Compared to continuous reactors, batch reactors are simple and easy to maintain and operate, and thereby have lower capital and operating costs. However, batch reactors only produce significant amounts of biogas during their peak production days, which limits the methane productivity. Also, an even and effective inoculation is hard to achieve in large-scale operation, or it requires high energy input for initial mixing, as discussed in section 3.2. In contrast, in continuous reactors the daily biogas production is stable after start-up, although additional agitation is required. Most of the studies on SS-AD of lignocellulosic biomass were conducted in batch reactors. Studies on biogas production from lignocellulosic biomass in continuous SS-AD reactors are needed. With data on the fundamental physicochemical and biological processes in SS-AD, further improvements of SS-AD reactor design can be proposed to enhance methane yield and reduce cost. More efforts should be devoted to innovative designs in inoculation, mixing, or leachate recirculation, which may solve the major limiting factors in SS-AD, such as the limited mass transfer, inhomogeneity, and accumulation of inhibitors, and facilitate the commercialization of SS-AD. To increase the loading of easily digestible feedstocks, two-stage systems should also be considered.

The translation of lab-scale SS-AD research to commercial digesters is another crucial aspect that needs more research efforts. Mass transfer, a key factor in SS-AD, is still under

investigation (Table 3). Heat transfer, another important aspect in scale-up, has rarely been studied in SS-AD. Modeling is a good option for these research areas, while more experimental studies are also required to obtain necessary data and verify model assumptions and results. Computational tools, such as computational fluid dynamics (CFD), that have been used in other relevant systems (e.g., soil, rumen, fermentation, L-AD, composting) can be employed in the scale-up and optimization of SS-AD. Techno-economic analyses are also necessary to select suitable reactor systems and improve the process efficiency and economic feasibility.

5.4. Assessment of digestate post-treatment technologies

Post-treatment of SS-AD finished material (digestate) is sometimes necessary to reduce moisture content, eliminate pathogens, and control the emission of residual biogas. Composting is one of the plausible technologies for post-treatment of digestate. Different from SS-AD, composting degrades organic matter by microorganism under aerobic conditions. Composting is simpler and cheaper than SS-AD, but does not produce biogas as a fuel (Li et al., 2011a; L. Lin et al., 2014). During the composting process, heat is generated due to aerobic microbial activities, which results in a high temperature that destroys pathogens. The compost can be used as a fertilizer or soil amendment. The high TS content of SS-AD digestate makes it suitable for treatment by composting, with only small amounts of additional bulking materials required. However, further research is needed to optimize the process and reduce the cost. Other post-treatment technologies are rarely studied for SS-AD, and future research should be conducted to discuss the impact of direct land application of digestate and the effects of different post-treatment methods.

5.5. Integration of SS-AD with other processes for value-added products

A common problem with SS-AD is the low value of the digestate, which could be addressed by integrating SS-AD with other processes to generate value-added products. One example is to integrate SS-AD of woody biomass with mushroom production (Y. Lin et al., 2015) (Figure 5). Sawdust or woodchips were used as the main substrate ingredient for Shiitake mushroom cultivation. About 60% of the cellulose and hemicellulose in the substrate were degraded during Shiitake cultivation (Y. Lin et al., 2015, 2014). However, a substantial amount of the lignin in the substrate was also decomposed and the substrate was permeated by mycelia during Shiitake growth, making the spent mushroom substrate (SMS) much more digestible than the untreated sawdust or woodchips (Y. Lin et al., 2015, 2014). According to Y. Lin et al. (2015), integrated Shiitake cultivation/SS-AD process improved the methane yield and solid reduction of woodchips by 1.5 and 8 times, respectively, compared with SS-AD of untreated woodchips (Figure 5). In addition, it was determined that for every kilogram of woodchips used, up to 89 g of Shiitake mushrooms could be produced as a high-value product (Y. Lin et al., 2015) (Figure 5). As a result, the integration of SS-AD with Shiitake cultivation is economically feasible. Besides, previous studies have also proposed to use SS-AD as a pretreatment method to produce bedding materials and particleboard (Zheng et al., 2009), or to use digestate as a substrate for bacteria and fungi to produce ethanol and lipids (Teater et al., 2011; Zhong et al., 2015). More efforts should be devoted to exploring the potential of SS-AD in integrated systems to generate fuels, chemicals, and other value-added products.

6. Conclusion

Compared to other lignocellulosic feedstocks, crop residues have higher methane production potential with SS-AD. Strategies, such as co-digestion, pretreatment, and optimization of operating conditions, are promising for improved methane yields and stability during SS-AD. Better understanding of mechanisms, such as mass transfer, biomass structure, and microbial distribution, during SS-AD has been achieved using modeling, microbial community analysis, and microscope imaging and tracer techniques. Reactor development is the limiting factor for commercialization of SS-AD, and future studies on other aspects, such as feedstock identification and co-digestion, feedstock storage/pretreatment, digestate treatment, and value-added production should also be conducted.

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Captions for Figures and Tables

Tables

Table 1 Potential of methane production by SS-AD of selected feedstocks in the U.S.

Table 2 Methane production by solid-state co-digestion of lignocellulosic biomass with other feedstocks

Table 3 Hypotheses of how TS content affects SS-AD process

Table 4 Recent mathematical models for SS-AD. Adapted from Xu et al. (2015)

Figures

Figure 1. Number of publications in which “solid state anaerobic digestion” appeared, based on search results via Google Scholar

Figure 2. Effect of pretreatment method and feedstock on methane yields during SS-AD. Adapted from Yang et al. (2015)

Figure 3. SS-AD digesters prepared with different premixing methods. Adapted from Zhu et al. (2015) (a): complete premixing, (b): partial premixing with the remaining inoculum in one layer, (c): partial premixing with the remaining inoculum in two layers.

Figure 4. Overall perspectives for further research in SS-AD of lignocellulosic biomass

Figure 5. Integrated Shiitake cultivation and SS-AD process for utilization of woody biomass. Adapted from Y. Lin et al. (2015)

Table 1 Potential of methane production by SS-AD of selected feedstocks in the U.S.

Feedstocks	CH ₄ yield ^a (m ³ /tonne VS)	VS (%, TS based)	Production (million tonnes/year)	CH ₄ potential ^b (million m ³ /year)
Crop residues				
Corn stover	239 ^[1]	94.9 ^[1]	75.0 ^[2]	17,010
Wheat straw	200 ^[3]	92.7 ^[3]	33.0 ^[4]	6,121
Municipal solid waste and forestry waste				
Yard trimmings	74 ^[3]	94.7 ^[3]	34.0 ^[5]	2,383
Forestry hardwood residues	47 ^[6]	98.5 ^[6]	30.7 ^[7]	1,420
Forestry softwood residues	17 ^[6]	97.3 ^[6]	15.6 ^[7]	259
Energy crops				
Giant reed	147 ^[8]	92.2 ^[8]	10.0 ^c	1,354
Miscanthus	130 ^[9]	96.8 ^[9]	4.9 ^c	617
Switchgrass	117 ^[6]	96.7 ^[6]	3.3 ^c	377

a: Cumulative CH₄ yield for 30-day SS-AD at 37°C;

b: CH₄ potential was calculated as: CH₄ yield (m³/tonne VS) × VS (%) × Production (million tonnes/year);

c: Production potential of each energy crop was calculated by its reported yield (45, 22.1 and 15 tonnes/ha/year for giant reed, miscanthus, and switchgrass, respectively) (Corno et al., 2014), and the assumption of using 20% of marginal lands (221,963 ha) in U.S. (Izaurrealde and Zhang, 2013);

[1]: Xu et al., 2013; [2]: Ertl, 2013; [3]: Y. Lin et al., 2014; [4]: Cardona et al., 2009; [5]: USEPA, 2015; [6]: Brown et al., 2012; [7]: Walsh, 2008; [8]: Liu et al., 2015a; [9]: Vasco-Correa and Li, 2015.

Table 2 Methane production by solid-state co-digestion of lignocellulosic biomass with other feedstocks

Feedstocks	Mixing ratio ^c	TS (%) / T (°C)	C/N	Increase (%)	References
Corn stover/expired dog food	1:1	22/37	11	129	Xu and Li, 2012
Rice straw/paper mill sludge	1:2 ^a	20/35	-	691 ^b	Mussoline et al., 2013
Rice straw/paper mill sludge/pig wastewater	1:1:1 ^a	20/35	-	679 ^b	Mussoline et al., 2013
Yard waste/food waste	4:1	19/36	<17	130	Brown and Li, 2013
Hay/soybean processing waste	1:1.3	18/37	12	148	Zhu et al., 2014
Yard trimmings/spent mushroom substrate	1:1	20/37	21	100	Y. Lin et al., 2014
Wood chips/ lawn grass/maple leaves	1:1:1	22/55	14	80-200	L. Lin et al., 2015

a: wet weight based; b: compared to the methane yield of rice straw; c: based on VS

Table 3 Hypotheses of how TS content affects SS-AD process

Feedstock and TS range	Proposed mechanism	References
Cardboard, TS%: 10%-35%	Limited liquid-gas mass transfer caused H ₂ and CO ₂ accumulation at high TS content and inhibited methanogens	Abbassi-Guendouz et al., 2012; Cazier et al., 2015
Corn stover, TS: 1%-28%	Diffusion limitation at high TS content caused sugar accumulation and inhibited the hydrolysis of substrates	Xu et al., 2014b
Rice straw and food waste, TS: 4.5%-23%; MSW, TS: 18%-25%;	Decreased mass transfer at high TS content reduced the substrate accessibility to microbes	Liotta et al., 2015; Pommier et al., 2007
Rice straw & food waste, 4.5%-23%	Decreased disintegration coefficient and hydrolysis rates at high TS content	Liotta et al., 2014
Wheat straw, TS: 10%-33%	AD metabolic pathway was changed with TS	Motte et al., 2013b
Acetate, propionate and cellulose, TS: 18%-35%	Reduced water availability to microbes at high TS content reduced microbial metabolic activity	Bollon et al., 2011; Le Hyaric et al., 2012, 2011

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823 Table 4 Recent mathematical models for SS-AD. Adapted from Xu et al. (2015)

	Category	Models	Features/Assumptions	Findings
1.	Modified ADM1	Theoretical	Based on ADM1 assuming homogenous system with modification of key parameters to be TS dependent.	Some mass transfer or microbial growth related kinetic coefficients in L-AD need to be adjusted with TS content to fit SS-AD data.
2.	Diffusion limitation	Theoretical	Diffusion limitation causes sugar accumulation, which inhibits the hydrolysis of substrates. An effective diffusion co-efficient in SS-AD is TS dependent.	Reaction rate increases with TS following Monod kinetics until a threshold is reached, and then decreases due to diffusion limitation.
3.	• Logistic • General kinetic	Empirical	Correlates key parameters of empirical models with operating conditions, such as TS content, and hydrolytic retention time.	The maximal specific growth rate of microbes and substrate accessibility to microbes are related to TS content.
4.	• Linear regression • ANN	Statistical	Uses extensive experimental data for model development/calibration/validation.	Both substrate characteristics and operating parameters affect SS-AD. Operating parameters have complicated relationships with reactor performance.

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Figure 1. Number of publications in which “solid state anaerobic digestion” appeared, based on search results via Google Scholar

(a)

(b)

Figure 2. Effect of pretreatment method and feedstock on methane yields during SS-AD.

Adapted from Yang et al. (2015)

Figure 3. SS-AD digesters prepared with different premixing methods. Adapted from Zhu et al. (2015)

(a): complete premixing, (b): partial premixing with the remaining inoculum in one layer, (c): partial premixing with the remaining inoculum in two layers.

Figure 4. Overall perspectives for further research in SS-AD of lignocellulosic biomass

Figure 5. Integrated Shiitake cultivation and SS-AD process for utilization of woody biomass.

Adapted from Y. Lin et al. (2015)